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14. ABSTRACT We have produced large sized 3D magnetic nanocomposites using a current activated pressure assisted densification (CAPAD). We were successful in producing dense nanocrystalline (< 100 nm average grain size) materials based on iron oxides (ferrites), rare earth oxides and silicon. We have demonstrated magnetic coupling leading to exchange bias in large nanocrystalline iron oxides. Additionally we can orient the magnetically coupled grains causing their magnetic properties to be highly anisotropic. These materials have promising applications in magneto-resistance based devices as well as permanent magnets. The rare earth oxides are transparent to visible light and cause very high Faraday rotations. The Verdet constant of these nanocrystalline materials is more than twice that of the state of the art Faraday rotation materials. We also used the CAPAD technique to show that the thermal conductivity in of polycrystalline silicon can be controlled using nanostructure.					
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## **(YIP 09) Producing Three Dimensional Nanostructured Magnetic Materials for Novel Magnetic Devices**

**Grant/Contract #:** FA9550-09-1-0197

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### **Abstract**

We have produced large sized 3D magnetic nanocomposites using a current activated pressure assisted densification (CAPAD). We were successful in producing dense nanocrystalline ( $< 100$  nm average grain size) materials based on iron oxides (ferrites), rare earth oxides and silicon. We have demonstrated magnetic coupling leading to exchange bias in large nanocrystalline iron oxides. Additionally we can orient the magnetically coupled grains causing their magnetic properties to be highly anisotropic. These materials have promising applications in magneto-resistance based devices as well as permanent magnets. The rare earth oxides are transparent to visible light and cause very high Faraday rotations. The Verdet constant of these nanocrystalline materials is more than twice that of the state of the art Faraday rotation materials. We also used the CAPAD technique to show that the thermal conductivity in of polycrystalline silicon can be controlled using nanostructure. These results are discussed in more detail below:

#### **(a) Exchange bias in large three dimensional iron oxide nanocomposites**

We have developed a processing method is presented for the production of macroscopic nanocomposites that display antiferromagnetic/ferrimagnetic (AFM/fM) coupling. The technique takes advantage of the metastability of iron oxide phases and the fast densification of nanocrystalline powders. The total processing time is under 500s. It is possible to manipulate the composition of fM and AFM phases with processing temperature. The relatively high density of AFM/fM boundaries produces an exchange bias caused by coupling at the interfaces. The magnitude of the exchange field  $H_{ex}$  is affected the composition as well as the grain size; the smaller grain size samples have the highest  $H_{ex}$ . This work was reported in *Applied Physics Letters* [1]

#### **(b) developemmet of CAPAD process for development of nanocryallinbe materissl**

Our technique is ideal for producing nanocomposite magnets because it affords: 1 ) Excellent control of grain size in the nanocrystalline regime 2) Ability to simultaneously densify different nanomagnetic material classes, *i.e.* produce large sized intimately mixed nanocomposites 3) Ability to achieve high quality interfaces such that exchange coupling can be achieved. 4) Ability to control the orientation of the magnetic phases so as to optimize coercivity ( $H_c$ ) and remanance magnetization ( $M_r$ ). A review of our processing technique was published in *Annual Review of Materials Research* [2].

### **(c) Magnetic and magneto-transport properties of highly anisotropic nanocrystalline ferrites**

In magnetic polycrystalline bulk materials, a crystallographically disordered microstructure produces directionally averaged magnetic properties. On the other hand, an ordered microstructure with aligned crystallographic magnetic directions can have higher coercivities, remanences, and/or exchange coupling. We produced preferentially ordered magnetic iron oxide composites using CAPAD. The morphology of the iron oxide powder crystallites was instrumental in creating bulk, magnetic composites with preferentially ordered microstructures. These magnetic composites possessed unequal (anisotropic) magnetic and magnetoresistive properties relative to the orientation of the samples to the application of an applied magnetic field. We plan to submit this work to *Physical Review B* [4].

### **(d) Producing Magneto-optical Nanocrystalline Oxides that display large Faraday Rotations**

Magneto-optical materials have widespread applications in communication and optical devices. Besides existing applications such as optical diodes, untapped potential applications could be accessed should magneto-optical properties be improved such that smaller magnetic fields can be employed. Here we present an efficient method for fabricating oxide materials that possess excellent optical and magnetic properties—they are transparent to visible light yet have high magnetic susceptibility. Combined, these properties produce large Faraday rotations; the measured Verdet constant is  $> -300 \text{ rad T}^{-1}\text{m}^{-1}$  at 632.8 nm, a high value for a thick, optically transparent material. Because this Verdet constant is more than twice that of the state of the art material, these nanocrystalline oxides produce polarized light rotations with less than half the applied magnetic field necessary. They are made by densifying rare earth nanocrystalline powder into dense, large-sized bodies using CAPAD. The processing temperature is optimized in order to achieve sufficient density without causing excessive phase changes that would destroy light transparency. This process produces materials quickly ( $<20 \text{ min}$ ), which, combined with high magneto-optical properties, promises less expensive, smaller, more portable magneto-optical devices. These results were published in *Journal of Applied Physics* [5] and resulted in a patent application [P1]

### **(e) Controlling thermal transport through nanostructure**

The thermal conductivity reduction due to grain boundary scattering is widely interpreted using a scattering length assumed equal to the grain size and independent of the phonon frequency (gray). To assess these assumptions and decouple the contributions of porosity and grain size, five samples of undoped nanocrystalline silicon have been measured with average grain sizes ranging from 550 to 64 nm and porosities from 17% to less than 1%, at temperatures from 310 to 16 K. The samples were prepared using current activated, pressure assisted densification (CAPAD). At low temperature the thermal conductivities of all samples show a  $T^2$  dependence

which cannot be explained by any traditional gray model. The measurements are explained over the entire temperature range by a new frequency-dependent model in which the mean free path for grain boundary scattering is inversely proportional to the phonon frequency, which is shown to be consistent with asymptotic analysis of atomistic simulations from the literature. In all cases the recommended boundary scattering length is smaller than the average grain size. These results should prove useful for the integration of nanocrystalline materials in devices such as advanced thermoelectrics. This work was published in *Nano Letters* [6]

## **Patents**

[P1] Filed 09/1/2010: MAGNETO-OPTIC NANOCRYSTALLINE OXIDES AND METHODS OF FORMING THE SAME” J.E. Garay and J.R Morales

## **Papers**

[1] J. R. Morales, S. Tanju, W. P. Beyermann and J. E. Garay, “Exchange bias in large three dimensional iron oxide nanocomposites” *Applied Physics Letters* (2010), 96, 013102.

[2] J. E. Garay, “Current Activated Pressure Assisted Densification of Materials,” *Annual Reviews of Materials Research* (2010), 40, 445-468.

[3] J. R. Morales, J. E. Alaniz and J. E. Garay, “The Current Activated Pressure Assisted Densification technique for producing Nanocrystalline Materials,” *JOM* (2010), 62, 58-62.

[4] J. R. Morales, Y. Koderá and J.E. Garay, “Magnetic and magneto-transport properties of highly anisotropic nanocrystalline ferrites.” To be submitted to *Physical Review B*.

[5] J. R. Morales, N. Amos, S. Khizroev and J. E. Garay, “Magneto-optical Faraday Effect in Nanocrystalline Oxides,” *Journal of Applied Physics* (2011) 109, 093110.

[6] Z. Wang, J. E. Alaniz, W. Jang, J.E. Garay and C. Dames, “Thermal Conductivity of Nanocrystalline Silicon: Importance of Grain Size and Frequency-Dependent Mean Free Paths” *Nano Letters* (2011) 11, 2206-2213